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Gamma and neutron dose measurements with solid state nuclear track detectors

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Abstract : An attempt has been made to explore the possibility of using solid state nuclear track detectors for the estimation of gamma and neutron doses based on the use of changes in activation energy of degradation of these detectors, due to irradiation (gamma/neutron), as a means of dosimetry. Thermogravimetry (TG) has been applied as a tool for these studies carried out on Lexan and CR-39 track detectors. A linear relationship observed between the decrease in activation energy and the dose (gamma/neutron) received by the detectors suggests the possibility of the use of these detectors as gamma and neutron dosimeters.

Keywords : Gamma dose, neutron dose, Lexan, CR-39, activation energy, thermogravimetry.

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1. Introduction

Measurements of gamma and neutron doses are needed very often. Day and Stein [1] have suggested the use of changes in optical density of plastics, due to irradiation as a means of dosimetry. Such changes in optical density have been observed for polycarbonate films [2] due to gamma irradiation. Frank and Benton [3] have also investigated Lexan polycarbonate plastic and found that this can be utilized in measuring absorbed gamma doses. Neutron fluxes can be measured by B^{10} (n,α) Li^7 nuclear reaction and subsequently registering the α and Li^7 recoils in CN detector or by using U^{235} (n,f) nuclear reaction. Since the thermal degradation temperatures of plastics like polycarbonates [4,5] are directly influenced by the gamma dose, an attempt has been made in the present investigation to estimate gamma and neutron doses based on the use of changes in the activation energy of degradation of plastics due to irradiation as a means of dosimetry. Thermogravimetry (TG) has been applied as a tool for these studies carried out on Lexan and CR-39 track detectors. A linear relationship observed between the decrease in activation energy and the dose (gamma/

neutron) received by the detectors suggests the possibility of the use of these detectors as gamma and neutron dosimeters.

2. Experimental

The Lexan polycarbonate film $(C_{16}H_{14}O_3)_n$ used in the present work had a thickness of ~75 μm and was obtained from General Electric, Schenectady, NY, USA. TG and DTA curves were recorded between 25–700°C in air on' an ULVAC themoanalyser. Samples weighing about 10 mg each were placed in the platinum sample holder and the thermal decomposition measurements were done at a heating rate of 6°C/min in flowing air atmosphere.

For gamma irradiation of the samples, the film was cut into small pieces of size about 2 cm^2 area and these were irradiated with ^{60}Co gamma rays for different doses in the range 2.5–15.5 Mrad.

CR-39 nuclear track detector used in the present studies was having the molecular formula $(C_{12}H_{18}O_7)_n$. The thermal studies of unirradiated and thermal neutron-irradiated films were carried out in an argon atmosphere using differential thermal analysis (DTA) and thermogravimetry (TG). The thermoanalytical data were recorded at the heating rate of 6°C/min upto 700°C in a ULVAC thermoanalyser using sintered Al_2O_3 as the reference material for DTA. The weights taken for TG experiments were about 25, 15, 11 and 10 mg for unirradiated, 12, 24 and 36 h irradiated samples, respectively. The irradiation of the detectors was carried out at the thermal column of the Apsara Reactor of BARC. The neutron flux of the thermal column was also experimentally determined using the method reported from our laboratory [6] and was found to be 4.1×10^6 neutrons/cm^2/s. The samples were irradiated at this neutron flux for 12–36 h. Hence, the values of neutron fluence (neutron flux × time of irradiation in seconds) for the 12, 24 and 36 h irradiated CR-39 samples in the present experiments were 1.75×10^{11}, 3.51×10^{11} and 5.26×10^{11} n/cm^2 respectively.

3. Results and discussion

The TG and DTA curves of unirradiated Lexan polycarbonate in air atmosphere are shown in Figure 1. Only the TG curves were analysed in the present studies. All TG curves show two steps degradation. Irradiation lowers the temperature at which the decomposition begins (T_i) for both the steps. The temperatures corresponding to complete decomposition (T_f) are also lowered in irradiated samples (Table 1). It is clear from the table that gamma dose results a substantial decrease in the T_i and T_f temperatures of irradiated Lexan films. This decrease in temperature due to gamma radiation may be because of scission of macromolecules resulting certain changes in various properties of the polycarbonate film. One such change is the formation of molecules of smaller length and increased reactivity which in turn lowers the T_i and T_f temperatures of the film.

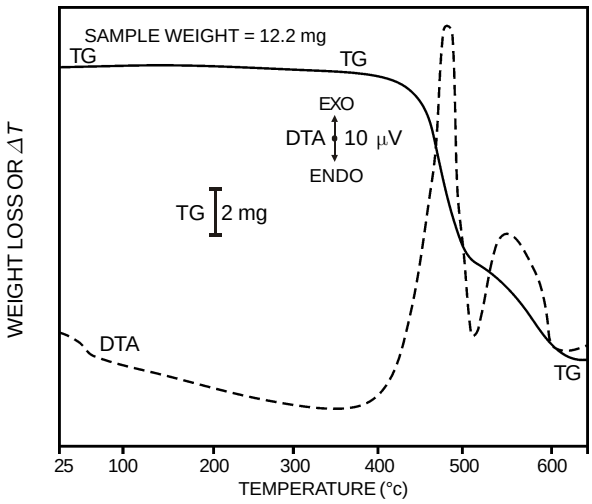

Figure 1. TG and DTA curves of Laxen polycarbonate.

Only the TG curves were analysed in the present studies. All the TG curves show two steps degradation. Irradiation lowers the temperature at which the decomposition begins (T_i) for both the steps. The temperatures corresponding to complete decomposition (T_f) are also lowered in the irradiated samples (Table 1). It is clear from the table that gamma dose results a substantial decrease in the T_i and T_f temperatures of irradiated Lexan films. This decrease in temperature due to gamma radiation may be because of scission of macromolecules resulting certain changes in various properties of the polycarbonate film. One such change is the formation of molecules of smaller length and increased reactivity which in turn lowers the T_i and T_f temperatures of the film.

Table 1. Thermal degradation data for unirradiated and gamma irradiated Lexan polycarbonate films.

Sample	TG temperatures		Wt loss/%	E/kJ mol
	T_i/°C	T_f/°C		
Unirradiated detector				
Step 1	330	510	66.4	327.0
Step 2	515	620	100.0	360.0
5.9 MRad				
Step 1	320	500	67.0	293.0
Step 2	505	605	100.0	317.0
10.0 MRad				
Step 1	315	500	60.0	273.0
Step 2	500	590	100.0	298.0
15.5 MRad				
Step 1	310	475	73.0	240.0
Step 2	490	570	100.0	262.0

In order to compare quantitatively the effects of gamma irradiation on the Lexan polycarbonate track detector, the kinetics of the two steps of thermal degradation were also investigated from the TG curves of the unirradiated and gamma irradiated samples according to the methods reported in the literature [7,8]. However our data for both unirradiated and irradiated Lexan polycarbonate track detector were found to fit well to Horowitz-Metzger method [5] although we had tried Freeman and Carroll and Coats and Redfern methods [7] also. In Horowitz-Metzger method $\log[\log(W_0/W)]$ is plotted *versus* θ where W_0 and W are the weights of the film initially and at time t, and $\theta = T - T_s$ where T is the temperature at weight W of the film and T_s is the temperature at which W_0/W equals $1/e$. The slope of the straight line thus obtained is given by $E/2.3RT_S^2$ where R is the gas constant and E is the energy of activation. The activation energy (E) values obtained by Horowitz-Metzger method from the kinetics studies for both the steps of degradation are presented in Table 1. The RSD (Relative standard deviation) obtained on E values by analysing few sets two times was found to be ±5%. It is obvious that gamma irradiation results in a substantial decrease in E in that values decrease with increasing dose. This decrease in activation energy values indicates that scission is predominant in gamma irradiation of Lexan polycarbonate track detector. Thus polycarbonate track detector undergoes scission upon gamma irradiation. A calibration curve drawn between T_f temperatures and the corresponding gamma doses was found to be linear. It is observed from the data presented in Table 1 that the plots of E *versus* dose in MRad also yield straight lines (Figure 2). It is felt that this plot is more representative of the processes taking place in the material

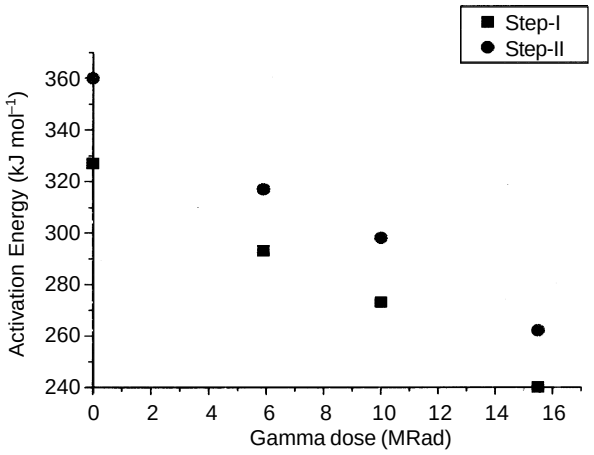

Figure 2. Plots of activation energy *vs.* gamma dose for Lexan polycarbonate.

as compared to the linear plot obtained between the temperature of completion of degradation and the gamma dose to which it is subjected. Further since the activation energy data is derived under fixed experimental conditions, the effect of gamma dose in it is highlighted in true sense. Moreover, it is directly connected with the mechanism of the process involved and would truly represent the influence of gamma dose on

scission. The quantities T_f, T_i *etc*. on the other hand are highly dependent on heating rate, sample size and other experimental parameters. This suggests the possibility of using the linearity between the E and gamma dose for the estimation of gamma dose.

The TG curves of CR-39 show that the unirradiated and the samples irradiated by thermal neutron for 12 h and 24 h degrade in two steps in argon atmosphere. The neutron irradiated samples for 36 h indicate only one stage of degradation in TG. The TG temperatures of inception for all these steps are given in Table 2.

Table 2. Thermal degradation data for unirradiated and thermal neutron irradiated CR-39 nuclear track detector films in an argon atmosphere.

Sample	TG temperatures		Wt. Loss/%	E/kJ mol
	T_i/°C	T_f/°C		
Unirradiated detector				
Step 1	200	450	87.5	102.0
Step 2	460	600	94.5	166.0
12 h thermal neutron irradiated detector				
Step 1	180	440	91.0	79.0
Step 2	450	550	98.0	134.0
24 h thermal neutron irradiated detector				
Step 1	170	385	75.0	64.0
Step 2	385	500	99.0	93.0
36 h thermal neutron irradiated detector				
Step 1	160	450	99.0	69.0

Irradiation lowers the temperature of inception (T_i) of the degradation steps. The final temperatures (T_f) of the degradation steps were also lowered in the neutron irradiated samples. In order to compare quantitatively the effects of thermal neutron irradiation on the CR-39 track detector, the kinetics of the different steps of thermal degradation were also investigated from the TG curves of the unirradiated and neutron irradiated samples according to the methods reported in the literature [7,8]. However, our data for both unirradiated and neutron irradiated track detectors were found to fit well to Horowitz-Metzger equation [5] rather than Freeman and Carroll and Coats and Redfern equations [7]. The activation energy (E) values obtained by Horowitz-Metzger method for all the steps of degradation were found to decrease with increase in neutron fluence (Table 2). It is observed from the data presented in Table 2 that the plots of E *versus* neutron fluence also yield straight lines (Figure 3).

This suggests the possibility of using the linearity between the E and neutron dose for the estimation of neutron dose. However, the two steps of degradation are poorly resolved in the case of 36 h irradiated sample. This is probably the reason for higher E-value in this case (average for the two steps) as compared to E-value for step

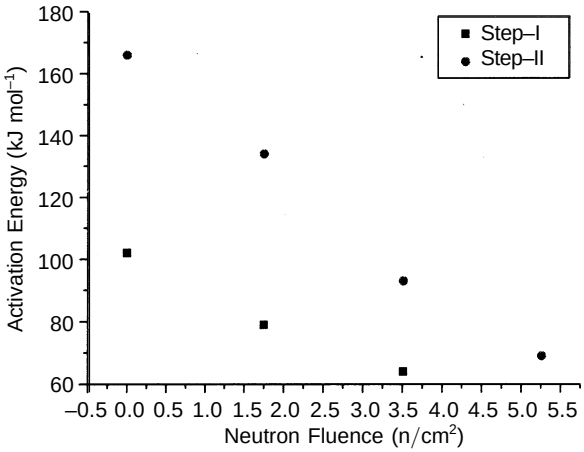

Figure 3. Plots of activation energy *vs.* neutron fluence for different stages of CR-39.

1 of 24 h irradiated sample. This decrease in activation energy values indicates that scission is predominant in thermal neutron irradiation of CR-39 track detector.

4. Conclusions

The results obtained in the present studies indicate the possibility of using Lexan polycarbonate and CR-39 films for the estimation of gamma and neutron dose based on the use of changes in the activation energy of degradation of plastics due to irradiation as a means of dosimetry.

Acknowledgments

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